Indian J. Phys. **73B** (1), 9-18 (1999)

IJP B
— an international journal

Seasonal distribution of CO_2, methane and water vapour over Delhi using solar IR spectroradio-meter

A B Ghosh†, S Bose† and R C Sharma*

†Radio and Atmospheric Sciences Division, National Physical Laboratory, Dr K S Krishnan Marg, New Delhi-110 012, India

*Dronacharya Government College, Gurgaon-122 001, Haryana, India

Received 29 April 1998, accepted 28 October 1998

Abstract An IR Spectro-radiometer has been installed at National Physical Laboratory New Delhi (28 7°, 77 2°E) India Several observations for the measurement of different atmospheric trace gases have been carried out The seasonal variation of atmospheric CO CH_4 and water vapour is presented in terms of integrated total column density An approach has been made to derive the surface concentrations from the integrated total columns Annual increases of CO, and CH_4 have been observed

Keywords CO_2, methane, water vapour solar infrared spectra

PACS No. 95 85 Hp

1. Introduction

Rapidly increasing human population and industrialisation have brought with them exotic man- made chemicals. This anthropogenic input of different gases is partly responsible for global warming of the earth's surface and atmosphere due to an enhancement of the greenhouse effect. The impact of the increase in greenhouse gases in the atmosphere on global climate is well established. The role of CO_2, CH_4 and water vapour in the discussion of global climate change is well recognised [1]. The release of carbon dioxide due to human activities has a significant increase in the atmospheric CO_2 content. Precise continuous measurements of the atmospheric CO_2 content have been performed since about 1958 at Mauna Loa, Howaii [2]. The increase of CO_2 in the atmosphere over the past century is expected to cause a warming of the earth. The surface concentrations around 315 ppm in 1958 are reaching ~360 ppm in 1997 [3,4]. The recent rate of increase in CO_2 has been sufficiently high enough to produce, if continued, an atmospheric burden before the middle of the next century of about 600 parts per million, approximately twice the amount thought to have been present in 1900 [5].

Methane is a very powerful greenhouse gas and is an active participant in chemical reactions in both the troposphere and stratosphere. It has a global warming potential (GWP)

that is over 20 times that of carbon dioxide (CO_2) on a molecule/molecule basis [6, 7]. The concentration has increased during the last 200 years [8,9] and accounts for approximately 15% of the anthropogenic greenhouse effects [10]. Lelieveld and Crutzen have estimated the climate forcing of methane for a 10 years time period to be 24.9 times that of CO_2, if indirect chemical effects are included. The measurement of methane levels in the atmosphere has shown on a general increase of 1% per year since the start of the industrial age [11]. This increase in CH_4 contributes not only to greenhouse effect, but also can lead to other atmospheric perturbations. An increase in tropospheric methane brings about an increase in tropospheric ozone levels and a decrease in tropospheric OH, the atmosphere's 'scrubber'. In the stratosphere, methane oxidation is an important source of water vapour and HO_x radicals, which have both important direct and indirect roles in stratospheric ozone depletion [12].

Water vapour is an important constituent in the atmosphere. It absorbs and emits infrared radiation and makes substantial contribution to the green house effect. In addition, water vapour molecules take part in the photochemical reactions above 60 km [13]. Accurate quantification of the greenhouse gases and their spatial and seasonal variation across the globe play an important role in understanding the atmospheric composition. The global community has begun to recognize that it is imperative to identify and monitor green house gases in both industrialized and developing countries. Several workers are monitoring different atmospheric gases throughout the globe. In India little work has been made in this direction so far. Thus, to quantify different atmospheric gases, a solar IR Spectro-radiometer has been installed at National Physical Laboratory, New Delhi (28.7°N, 77.2°E and 220 meter above sea level). Many spectra have been recorded to quantify the abundance of several trace species, including CO_2, CH_4 and water vapour. The seasonal variation in terms of total column amounts and the corresponding surface concentration have been retrieved from a set of solar IR spectra.

2. Experimental setup

A heliostat has been used for tracking the Sun. The solar radiation is focussed on the entrance slit of the spectro-radiometer by a newtonian telescope. The built-in chopper with a frequency

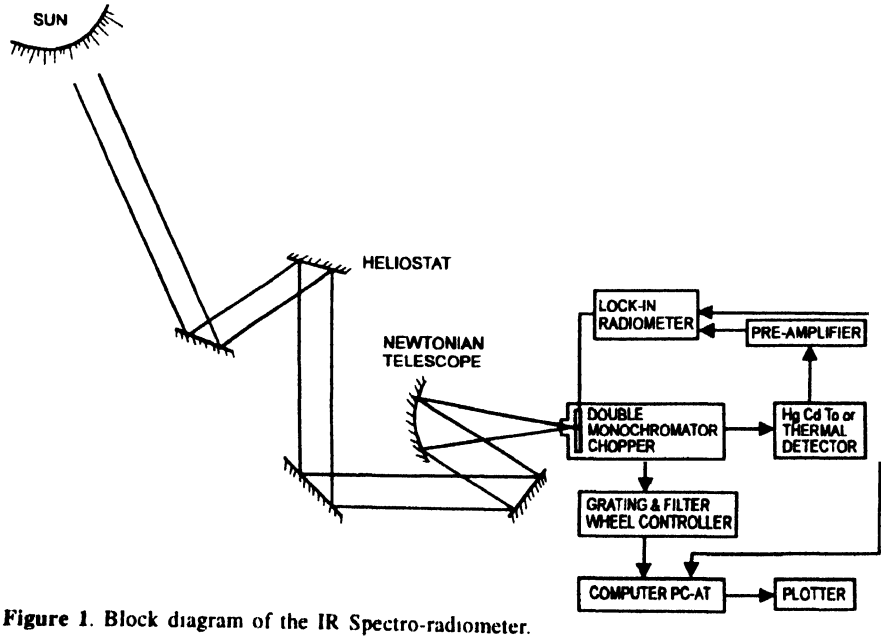

Figure 1. Block diagram of the IR Spectro-radiometer.

of 163.4 Hz at the entrance slit modulates the continuous light signal with a square wave. The incident radiation is dispersed by the gratings blazed at 1600 nm and passed through the optical band filter to the detector mounted at the exit slit. The electrical signal generated by the detector is amplified by the auto ranging lock-in amplifier. It has preset preamplifier and phase angle settings for four different detectors with a sensitivity of 10^{-14} ampere for the photovoltaic and of 10^{-7} volts for the photoconductive detectors. The output of the lock-in amplifier is fed to the computer for data processing and analysis. A block diagram of the system is shown in Figure 1. The experimental details of the system have been discussed in earlier papers [14, 15]. The system has been calibrated with the IR source and a wide range thermal detector with a KRS-5 window sensitive in the spectral range 700 nm to 20000 nm has been used for the present study. The calibration curve is shown in Figure 2.

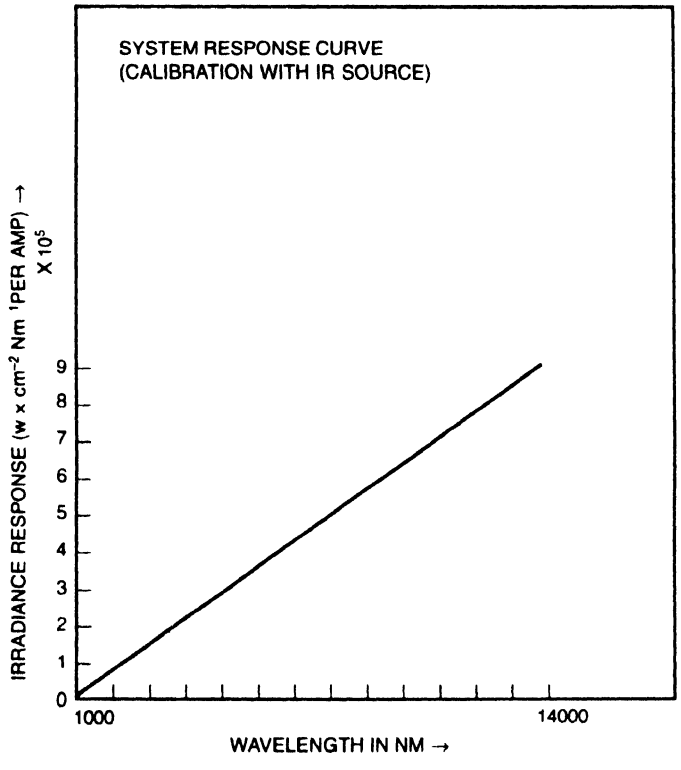

Figure 2 Calibration curve of the IR Spectro-radiometer

3. Observations and data analysis

Atmospheric CO_2, CH_4 and water vapour show characteristic absorptions in the IR spectral region. The spectral ranges 4375-4385, 7530-7540 and 6495-6500 nm have been used for CO_2, CH_4 and water vapour respectively. Direct solar spectra with a spectral resolution of 1 nm have been recorded on clear sky days. Examples of the observed spectra of CO_2, methane and water vapour are shown in Figure 3, 4 and 5 respectively.

The line strength and the Lorentz half width of the recorded spectrum has been deduced from the HITRAN 92 database [16]. The computation procedure for the integrated total column has been discussed earlier [14, 15].

The monthly averages of the integrated total columns have been arranged to illustrate the seasonal variation [Figures (6-12)]. The surface concentration of CO_2 and methane are deduced from the integrated total column. A model distribution of CO_2 and CH_4 is taken as

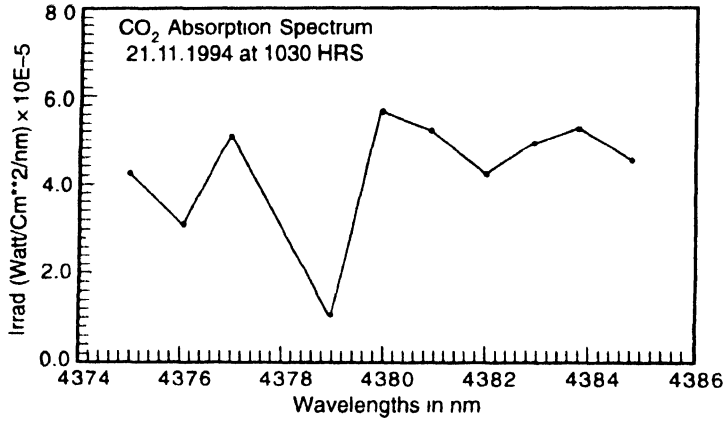

Figure 3 Observed CO_2 spectrum as recorded by the IR Spectro-radiometer

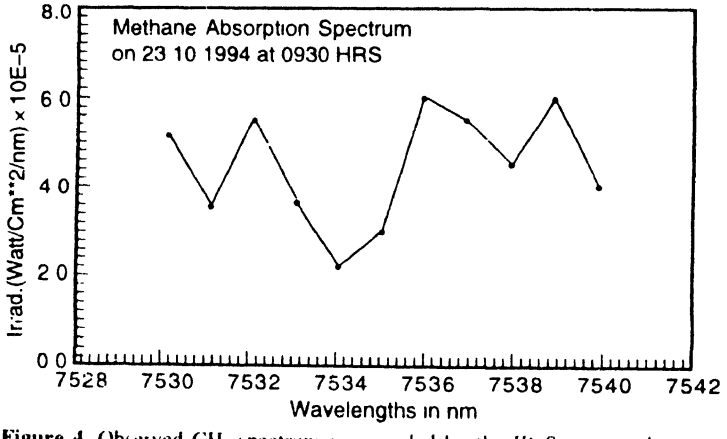

Figure 4 Observed CH_4 spectrum as recorded by the IR Spectro-radiometer

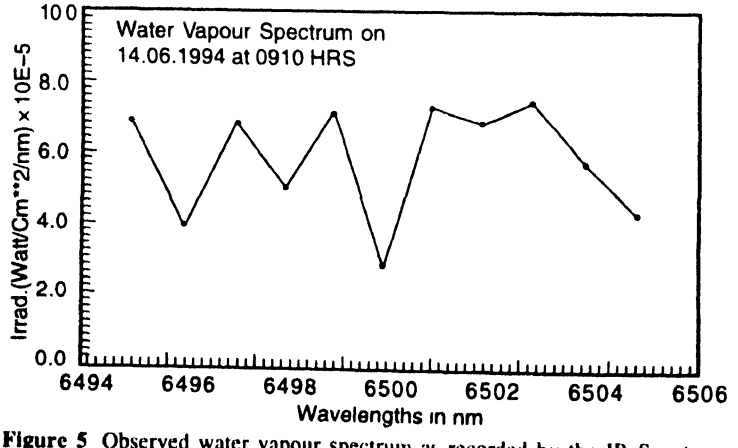

Figure 5 Observed water vapour spectrum as recorded by the IR Spectro-radiometer

initial guess [17]. The profile is modified so that after integration, the total column density matches with the measured column density of CO_2 and CH_4.

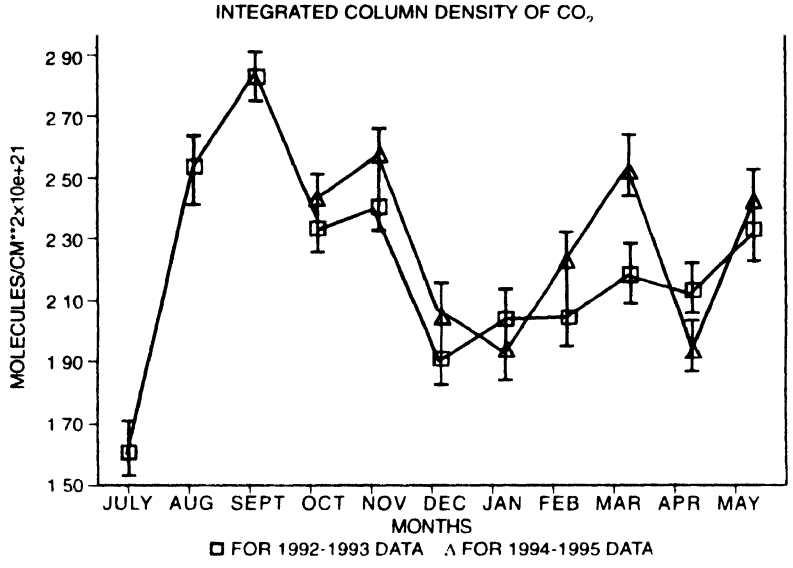

Figure 6 Monthly variation in total column amounts of CO_2 over Delhi

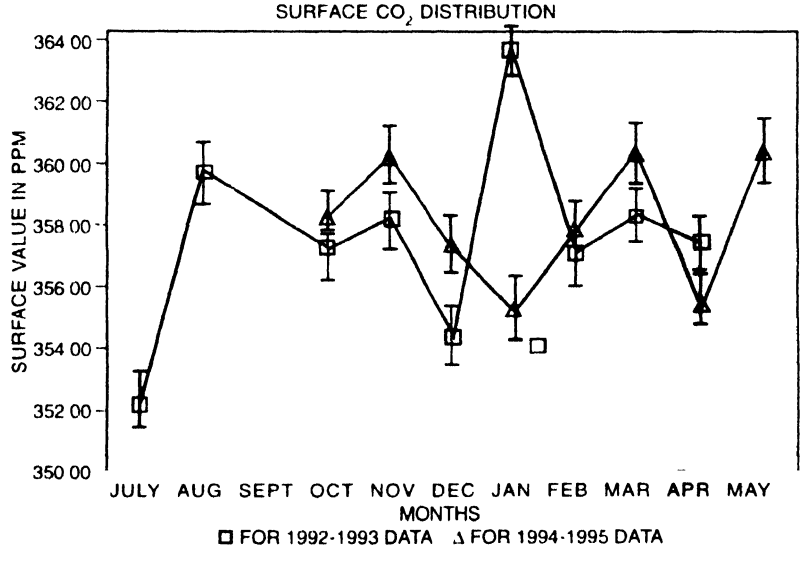

Figure 7 Deduced surface concentration of CO_2 over Delhi

4. Results and discussion

The monthly variations of atmospheric CO_2 for the years 1992-1993 and 1994-1995 are shown in Figure 6. The values vary between 1.58-2.87 molecules /cm^2 × 10^{21} for the year 1992-1993 and 2.00–2.60 molecules /cm^2 × 10^{21} in 1994-1995. The monthly means of CO_2 show a seasonal trend. Higher values are observed during the post-monsoon period and post-winter months and lower values in the winter months. The seasonal distribution of CO_2 may be affected due to the local and regional activities. With the onset of monsoon the concentration of CO_2 is found to be increasing as the rainy season progresses.

(iii) The uniqueness of the seasonal variation of CO_2 surface concentration at 3 global locations have maximum during winter and spring months.

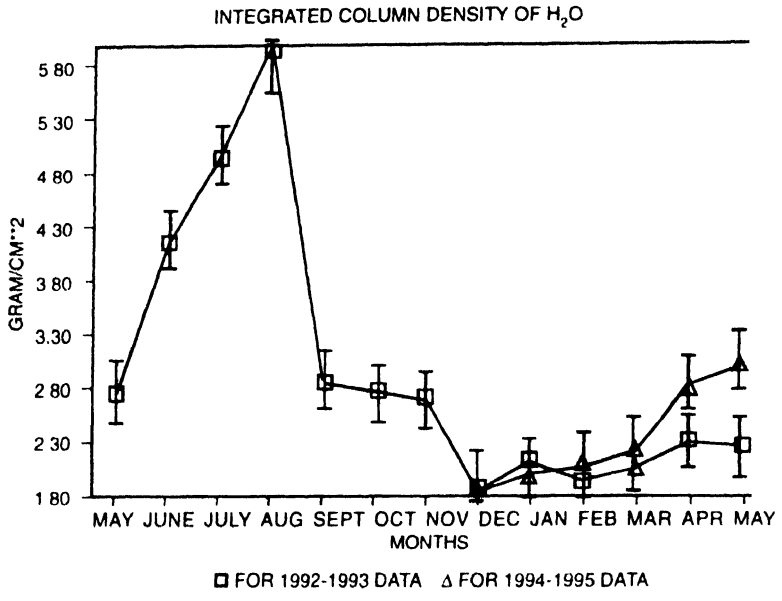

Figure 12 Monthly variation of water vapour column amount over Delhi

The monthly variations of atmospheric methane for the years 1992-1993 and 1994-1995 are shown in Figure 9. The monthly means of methane total column vary between 1 8-3.5 × 10^{19} molecules/cm^2 in 1992-1993 and 1.9- 3.3 × 10^{19} molecules/cm^2 in 1994-1995. The methane loads show a minimum in the winter months. This might be ascribed to the lower fermentation rate in the winter months. The observed higher values in August-Sept 1992 may have their cause in the decaying of different biological and other materials in the environment due to the rain of SW-monsoon. From Figure 9, it can be seen that methane increased from 1992-1993 to 1994-1995. This year wise increase may be due to the agricultural practices, cattle and dairy farming, rice growing, slash and burn agriculture, burning of fuel wood and decomposition of agricultural wastes. The increase in atmospheric methane is largely due to an increase in man made sources and is coupled with the increase in world population

The surface methane concentrations for the same years and months are shown in Figure 10. The values vary between 1700-2300 ppm in 1992-1993 and 1700-1950 ppm in 1994-1995. The higher concentration of CH_4 during the month of September may be due to the increase in the mixing layer height. Though a higher surface concentration is observed during December 1992, the yearly average shows an increase for the year 1994-1995. This increase is likely to be anthropogenic, *i.e.* caused by population growth, agriculture, energy use, industrial activities and transportation.

In Figure 11, a comparison is made between Delhi (28.7°N, 77.2°E, 220 m above MSL), Mauna Loa (19.3°N, 155.3°W, 3397 m above MSL) and Cape Meares (45.3°N, 120°W, 30 m above MSL) in Figure 11. The following information can be drawn from the comparison :

(i) The monthly variations of the CH_4 concentrations are different for Delhi, Mauna Loa and Cape Meares.

(ii) The concentrations of CH_4 at Delhi, Mauna Loa and Cape Meares agree within a range of 0.5 ppm in their monthly means except for September and March. The concentration of CH_4 at Delhi, India, are higher by 5 ppm during the months of September and March. The higher concentration of CH_4 during September at Delhi as compared to Mauna Loa or Cape Meares is due to a higher mixing length due to enhanced atmospheric turbulance.

The water vapour distribution over Delhi is shown in the Figure 12. The values vary between 1.94-6 gram/cm^2. The observed water vapour shows higher values in the months of July and August. This trend is due to the active SW-monsoon in this region.

5. Summary

The seasonal distribution of integrated total column density of CO_2 and methane and water vapour have been measured over Delhi using Solar IR spectro-radiometer. Based on the guess profile and the column density the surface concentration of CO_2 and CH_4 have been deduced over Delhi. Though the system has some limitations for the observations during the monsoon months and the surface concentrations are deduced based on the guess profile yet for a long term data base of the greenhouse gases and quantitative analysis over the Indian sub continent it provides an unique representation.

The monthly values of atmospheric CO_2 for the years 1992-1993 and 1994-1995 vary between 1.58-2.87 molecules/cm$^2 \times 10^{21}$ and 2.00-2.60 molecules/cm$^2 \times 10^{21}$ respectively. The monthly means of CO_2 show a seasonal trend, higher during the post-monsoon period and post-winter months and lower in the winter months The surface CO_2 concentrations for the years 1992-93 and 1994-95 vary between 353-364 ppm and 356-361 respectively. Higher concentration of CO_2 during September and October month have been observed and a year wise increasing trend for the year 1992-93 to 1994-1995 have been found.

The comparison of Delhi (28.7°N, 77.2°E, 220 m above MSL), Mauna Loa (19.3°N, 155.3°W, 3397 m above MSL) and Westerland (55°N, 8°E, 8 m above MSL) shows the concentrations of CO_2 at Delhi and Mauna Loa largely agree within a range of 5 ppm. However the concentration of CO_2 at Westerland, Germany are lower by 20 ppm. The uniqueness of this comparison is that the seasonal variation have maximum during spring months.

The monthly values of atmospheric methane for the years 1992-1993 and 1994-1995 vary between 1.8-3.5 $\times 10^{19}$ molecules/cm^2 and 1.9-3.3 $\times 10^{19}$ molecules/cm^2 respectively. The methane loads show a minimum in the winter months and higher values in August-Sept 1992. There is also a year wise increase of methane observed from 1992-1993 to 1994-1995. The surface methane concentrations for the same years and months vary between 1700-2300 ppm in 1992-1993 and 1700-1950 ppm in 1994-1995. Higher concentration for CH_4 has been observed during the month of September and the yearly average shows an increase for the year 1994-1995 than 1992-1993.

The comparison of Delhi (28.7°N, 77.2°E, 220 m above MSL), Mauna Loa (19.3°N, 155.3°W, 3397 m above MSL) and Cape Meares (45.3°N, 120°W, 30 m above MSL) agree within a range of 0.5 ppm in their monthly means except for September and March. The concentration of CH_4 at Delhi, India, are higher by 5 ppm during the months of September and March.

The water vapour values vary between 1.94-6 gram/cm^2. The observed water vapour shows higher values in the months of July and August.

References

[1] A M Thomson and R J Cicerone *J Geophys Res* **91** 10853 (1986)

[2] C D Keeling *Tellus* **25** 174 (1973)

[3] *Climate Change 1995*, eds J T Houghton *et al*. (Cambridge : Cambridge University Press) (1996)

[4] F S Rowland and I S A Isaksen *The Changing Atmosphere . Physical, Chemical and Earth Sciences* Research Report 7 (New York John Wiley & Sons) p 1 (1988)

[5] G M Woodwell, J E Hobbie, J M Melillo, B Moore, B J Peterson and G R Shaver *Science* 1081 (1983)

[6] IPCC, *Climate Change-The IPCC Scientific Assessment, Intergovernmental Panel on Climate Change*, (Cambridge Cambridge University Press) (1990)

[7] J Lelieveld and P J Crutzen *Nature* **355** 339 (1992)

[8] G I Pearman, D Etheridge, F DeSilra and P J Fraser *Nature* **320** 248 (1986)

[9] L P Steele, E J Dlugokency, P M Lang, P P Tans, R C Martin and K A Masarie *Nature* **358** 313 (1992)

[10] H Rodhe *Science*, 1217 (1990)

[11] D R Blake and F S Rowland *Science* **239** 1129 (1988)

[12] J H Shorter, J B MCmanus and C Kolb *et al J Atmos Chem*. **24** 121 (1996)

[13] J T Houghton and J S Seely *Quart J Roy Meteor Soc* **86** 358 (1960)

[14] A B Ghosh, R C Sharma and B M Reddy *Indian J Radio Space Phys* **22** 38 (1993)

[15] A B Ghosh, S Bose and R C Sharma *J Metrology Soc India*, **12** 17 (1997)

[16] L S Rothman, R R Gamache, R H Tipping, C P Rinsland, M A H Smith and R A Toth *The HITRAN Molecular Database Editions of 1991 and 1992, J Quant Spectrosc Radiat Transfer* **48** 469 (1992)

[17] A P Mitra *Minor Constituents in the Middle Atmosphere (Scientific Notes)* **ISRO-INCOSPAR-SN-03-77** (1997)

[18] T A Boden, D P Kaiser, R J Sepanski and F W Stoss *Trends '93 : A Compedium of Data on Global Change* **Pb. No. ORNL/CDIAC-65** (1994)

[19] L Ineborg, R Graul and N B A Trivett *Tellus* **47B** 23 (1995)